Development of High Efficiency Segmented Thermoelectric Couples for Space Applications

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Historical RTG-Powered U.S. Missions

Mission	RTG type (number)	TE	Destination	Launch Year	Mission Length	Power Level*
Transit 4A	SNAP-3B7(1)	PbTe	Earth Orbit	1961	15	2.7
Transit 4B	SNAP-3B8 (1)	PbTe	Earth Orbit	1962	9	2.7
Nimbus 3	SNAP-19 RTG (2)	PbTe	Earth Orbit	1969	> 2.5	~ 56
Apollo 12[#]	SNAP-27 RTG (1)	PbTe	Lunar Surface	1969	8	~ 70
Pioneer 10	SNAP-19 RTG (4)	PbTe	Outer Planets	1972	34	~ 160
Triad-01-1X	SNAP-9A (1)	PbTe	Earth Orbit	1972	15	~ 35
Pioneer 11	SNAP-19 RTG (4)	PbTe	Outer Planets	1973	35	~ 160
Viking 1	SNAP-19 RTG (2)	PbTe	Mars Surface	1975	> 6	~ 84
Viking 2	SNAP-19 RTG (2)	PbTe	Mars Surface	1975	> 4	~ 84
LES 8	MHW-RTG (2)	Si-Ge	Earth Orbit	1976	15	~ 308
LES 9	MHW-RTG (2)	Si-Ge	Earth Orbit	1976	15	~ 308
Voyager 1	MHW-RTG (3)	Si-Ge	Outer Planets	1977	40	~475
Voyager 2	MHW-RTG (3)	Si-Ge	Outer Planets	1977	40	~475
Galileo	GPHS-RTG (2)	Si-Ge	Outer Planets	1989	14	~ 574
Ulysses	GPHS-RTG (1)	Si-Ge	Outer Planets/Sun	1990	18	~ 283
Cassini	GPHS-RTG (3)	Si-Ge	Outer Planets	1997	20	~ 885
New Horizons	GPHS-RTG (1)	Si-Ge	Outer Planets	2005	12 (17)	~ 246
MSL	MMRTG (1)	PbTe	Mars Surface	2011	6 (to date)	~ 115
*Mars 2020***	*MMRTG (1 baselined)*	*PbTe*	*Mars Surface*	*2020*	*(5)*	*> 110*

[#]Apollo 12, 14, 15, 16 and 17
**Planned

**Total power at Beginning of Mission (W)*

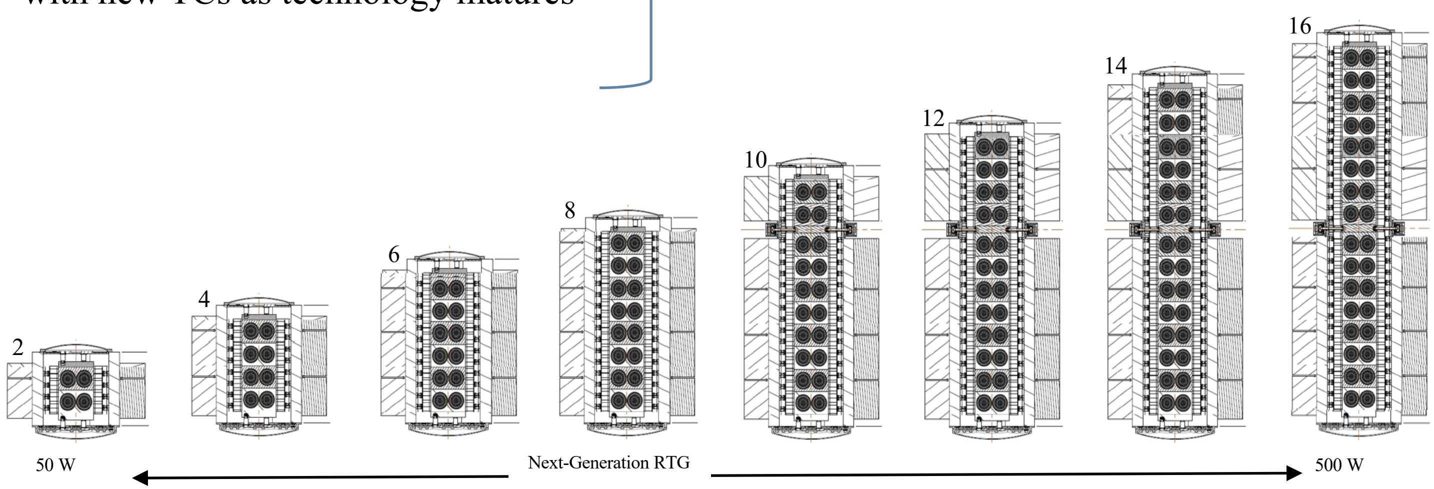
From a few watts up to ~ 900 W, up to 40 years of operation (and counting)

Next Generation RTG Study Recommendations

- **NG-RTG:**
  - Vacuum Only
  - Modular
- **Variants**: **2, 4, 6, 8, 10, 12, 14**, and **16 GPHS** variants
  - 16 GPHSs (largest RTG variant)
  - P_{BOM} = **400-500 W_e** (largest RTG variant)
  - Mass goal of < **60 kg** (largest RTG variant)
  - Degradation rate **< 1.9 %**
  - System to be designed to be upgraded with new TCs as technology matures

Three (3) couple configurations have been recommended, with conversion efficiencies ranging from *e=13%* to *e=16.4%*

Conversion efficiencies for heritage systems is e~ 7%



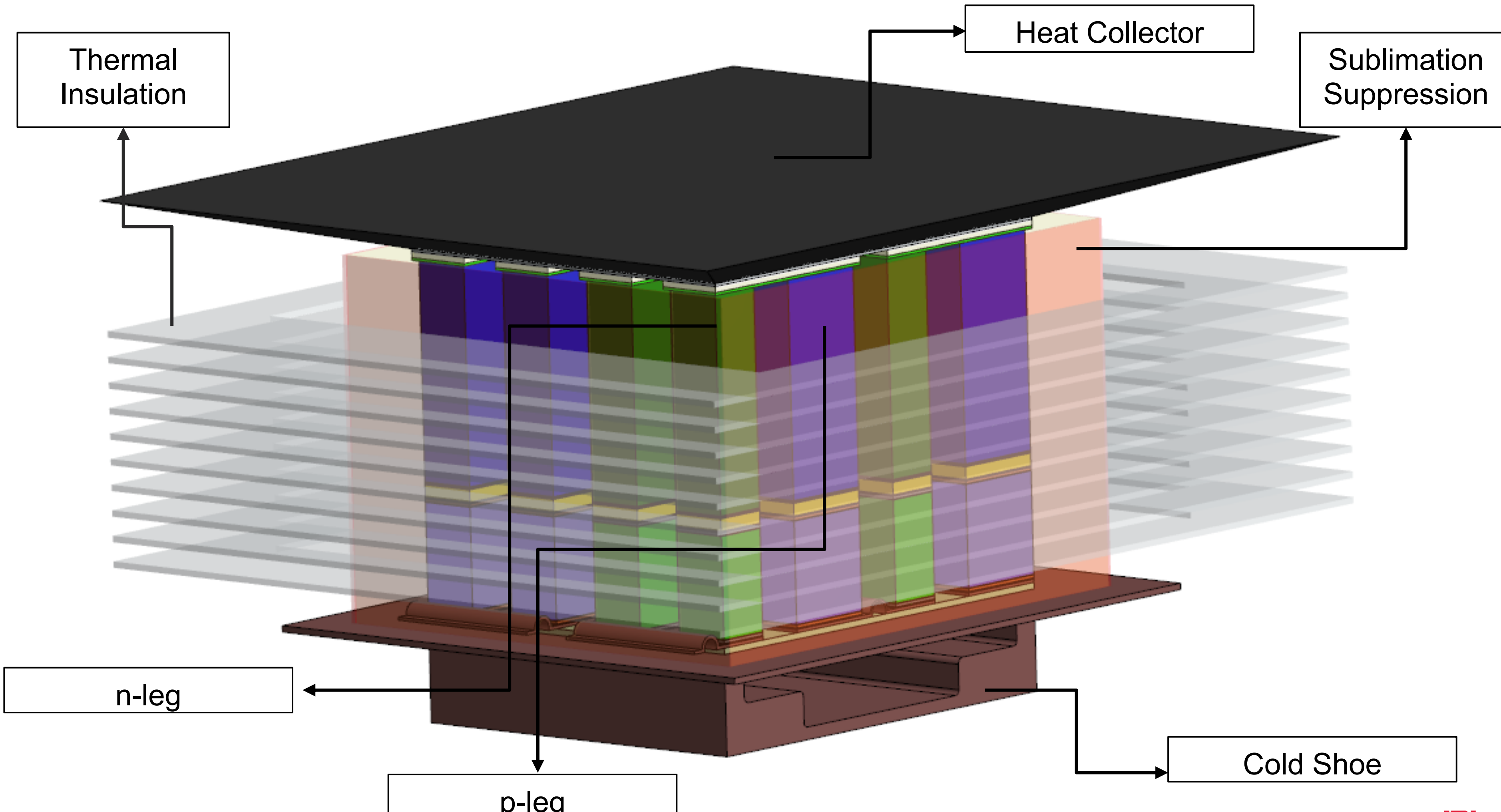
Candidate Couple Configurations

Configuration #	n		p		Current (study*) Predicted Materials-based Couple Efficiency (%)	Current (study*) Estimated BOL RTG Efficiency (%)	Current (study*) Estimated BOL RTG Power (W)
	Low	High	Low	High		*16-GPHS, 250W per GPHS*	
1	1-2-2 Zintl	$La_{3-x}Te_4$ /composite	9-4-9 Zintl	14-1-11 Zintl	**14.7** (16.4*)	**12.8** (14.3*)	**513** (572*)
3	SKD	$La_{3-x}Te_4$ /composite	SKD	14-1-11 Zintl	**13.9** (15.6*)	**12.1** (13.6*)	**485** (544*)
14		$La_{3-x}Te_4$ /composite		14-1-11 Zintl	**11.1** (13.0*)	**9.7** (11.3*)	**387** (452*)

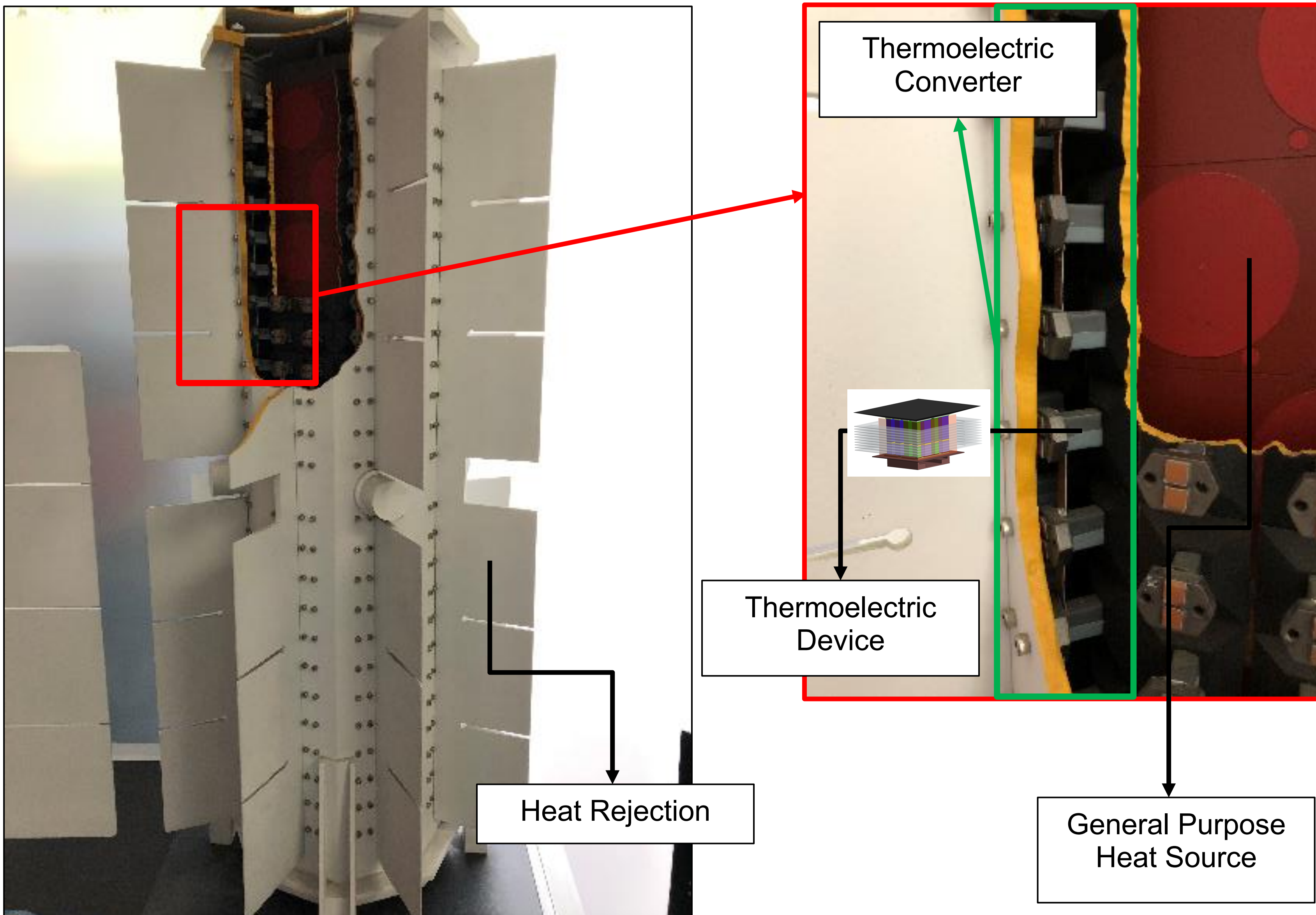
- Efficiency calculated during Next Gen RTG study used higher ZT $La_{3-x}Te_4$/composite material produced by small batch synthesis (15 gr).
  - Current Project baseline (April 2018) produced by large batch synthesis process (100 g)
  - **Process optimization to reproduce these original results is in progress**
- Couple efficiency based on couple operating $T_{hot\ junction}$ = 1273 K, $T_{inter\text{-}segment}$ = 773 K and $T_{cold\ junction}$ = 450K
  - **Lower temperature segments, such as SKDs, would operate no higher than 773 K (500 C)**
  - **Most of hot side interface degradation risk would be for 14-1-11 Zintl and La_{3-x}Te4/composite**
- Estimated BOL system-level efficiency based on heritage RTG performance (derating factor)
  - GPHS-RTG: couple (7.5%) ; system (6.5%)
  - MMRTG: couple (7.1%); system (6,3%)

Multicouple Device for Modular System Concept

- ≥ 11% system conversion efficiency (≥ 60% improvement over MMRTG at BOL)
- ≥ 6-8.5 We/kg specific power (2-3 x improvement over MMRTG)
- 1.9%/year or lower power degradation average over 17 years (including isotope decay)

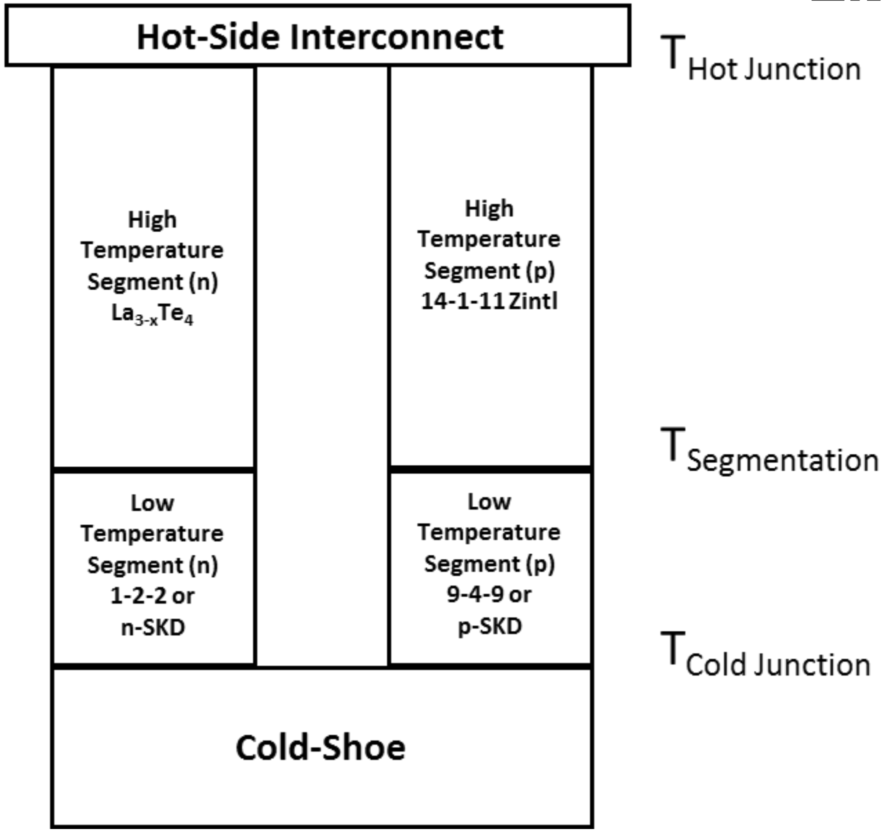


Radioisotope Thermoelectric Generator



Managing Technology Development Risk

Ex. All Zintl Segmented Couple



T_{HOT} (K)	T_{COLD} (K)	Efficiency* (%)
1273	450	15.8
1223	450	15.2
1173	450	14.5
1123	450	13.7
1073	450	13.0
1023	450	12.2
973	450	11.4

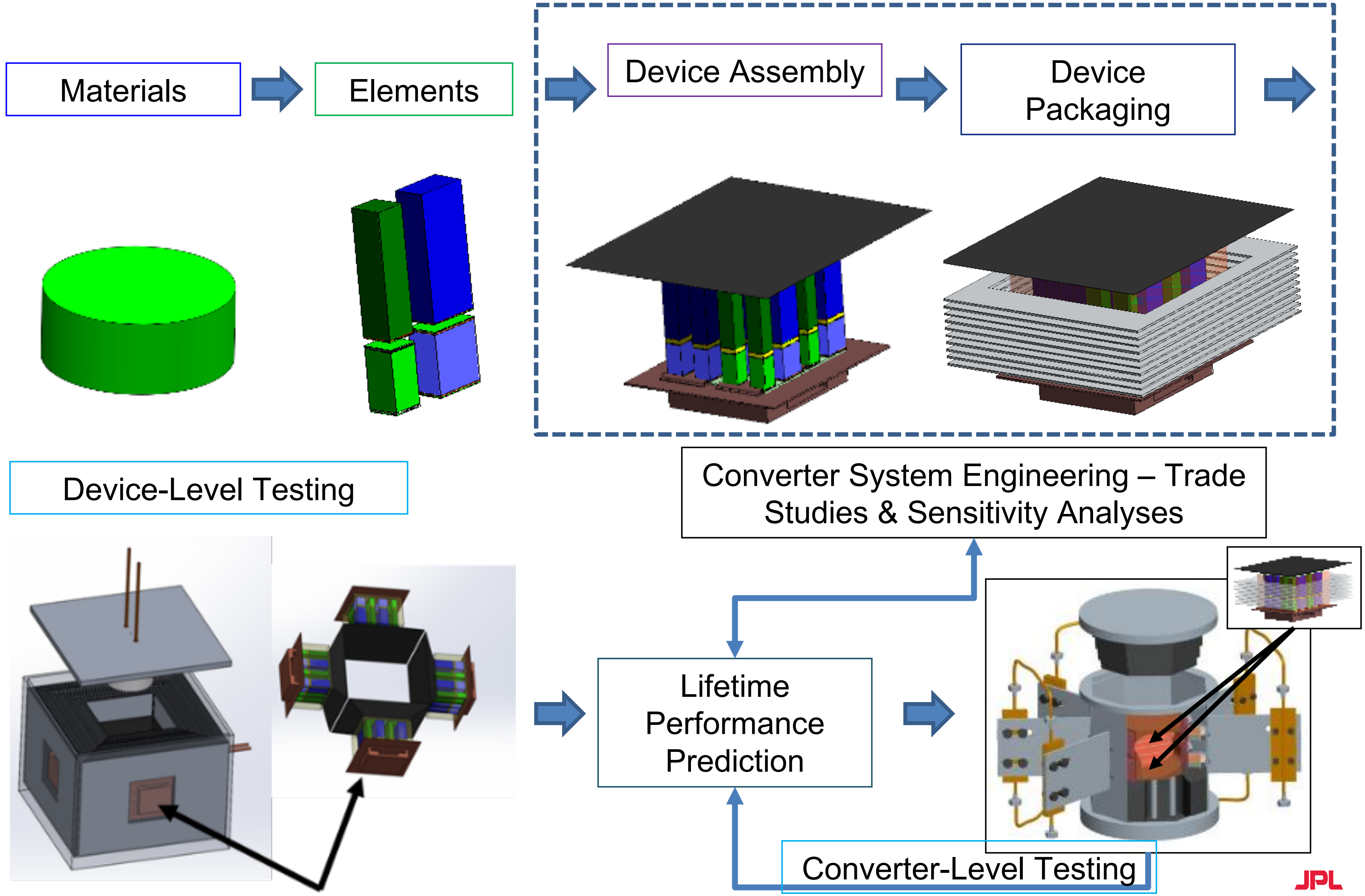
500W target (1223 K row)

400W target (1023 K row)

**Preliminary prediction at Beginning-of-Life (BOL) – will be updated periodically*

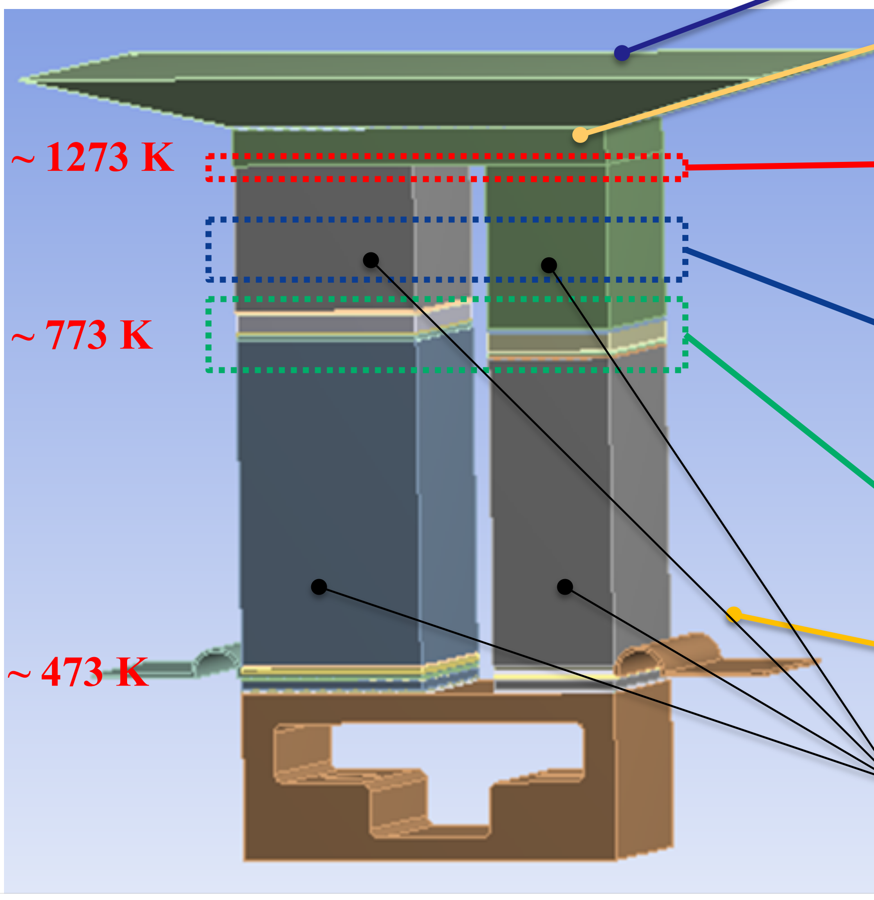
…We can reduce hot-junction temperature and subsequently minimize degradation rate

Converter Technology Development



Key Materials Challenges

- The temperatures of operation coupled with the device architecture (brittle intermetallic materials, metal/semiconductor contacts) give rise to a unique materials challenges:



Hot shoe with dielectric layer (multicouples)

Compatible interconnect material(s)

- Development of mechanically robust and stable hot-side interfaces

- Development of mechanically robust TE materials that display stable TE performance for thousands of hours and at temperature.

- Mechanically compliant, high electrical/thermal conduction inter-segment interfaces

- Aerogel thermal insulation / sublimation suppression / Chemical reactivity suppression

Scale-up synthesis of all TE Materials

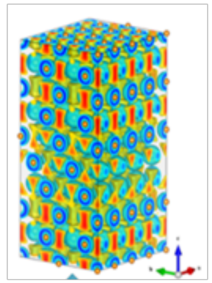
All Challenges must be addressed for the project to be successful!

Thermodynamic Phase Stability Calculations to Guide Materials Selection

- Develop database for TE materials of interest
- Test models against experimental data
- Use model to guide materials selection for developing stable, highly conductive interfaces and improved thermomechanical performance.

Crystal Structure

↓



Electronic structure

First-principles calculations based on DFT (via VASP)

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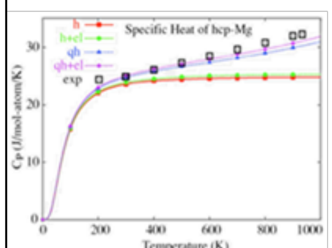

Thermochemical properties

Statistical Mechanics

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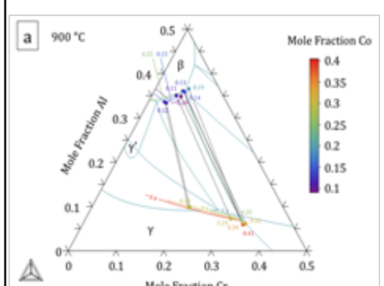

Phase Diagrams

CALPHAD Modeling

Kinetic Modeling

DICTRA Modeling

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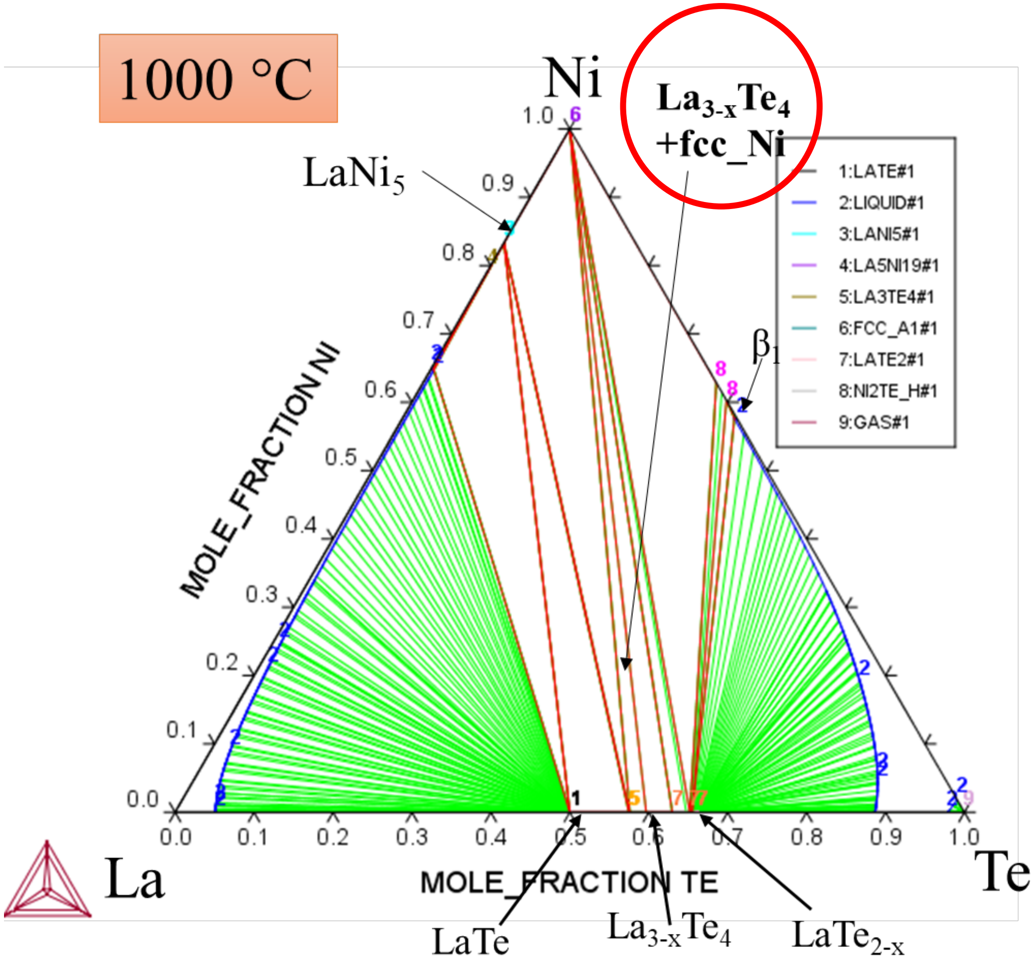
Example: Thermodynamic Phase Stability of Ni-$La_{3-x}Te_4$



- **Thermodynamic model predicts $La_{3-x}Te_4$ and Ni** exist in a stable two phase only region validating the long term stability of the two phases
- Thermodynamic modeling maybe used to select "compositing" approach for other relevant TE materials.
- Effect of compositing is the improvement of mechanical properties.

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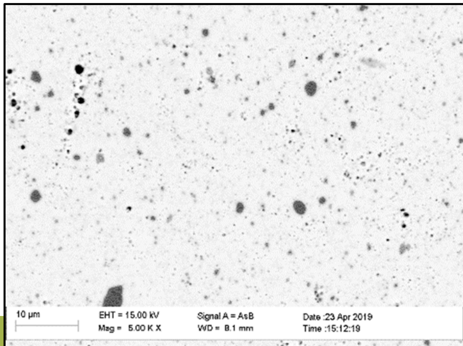
Mechanically Robust TE Material via Metal Compositing

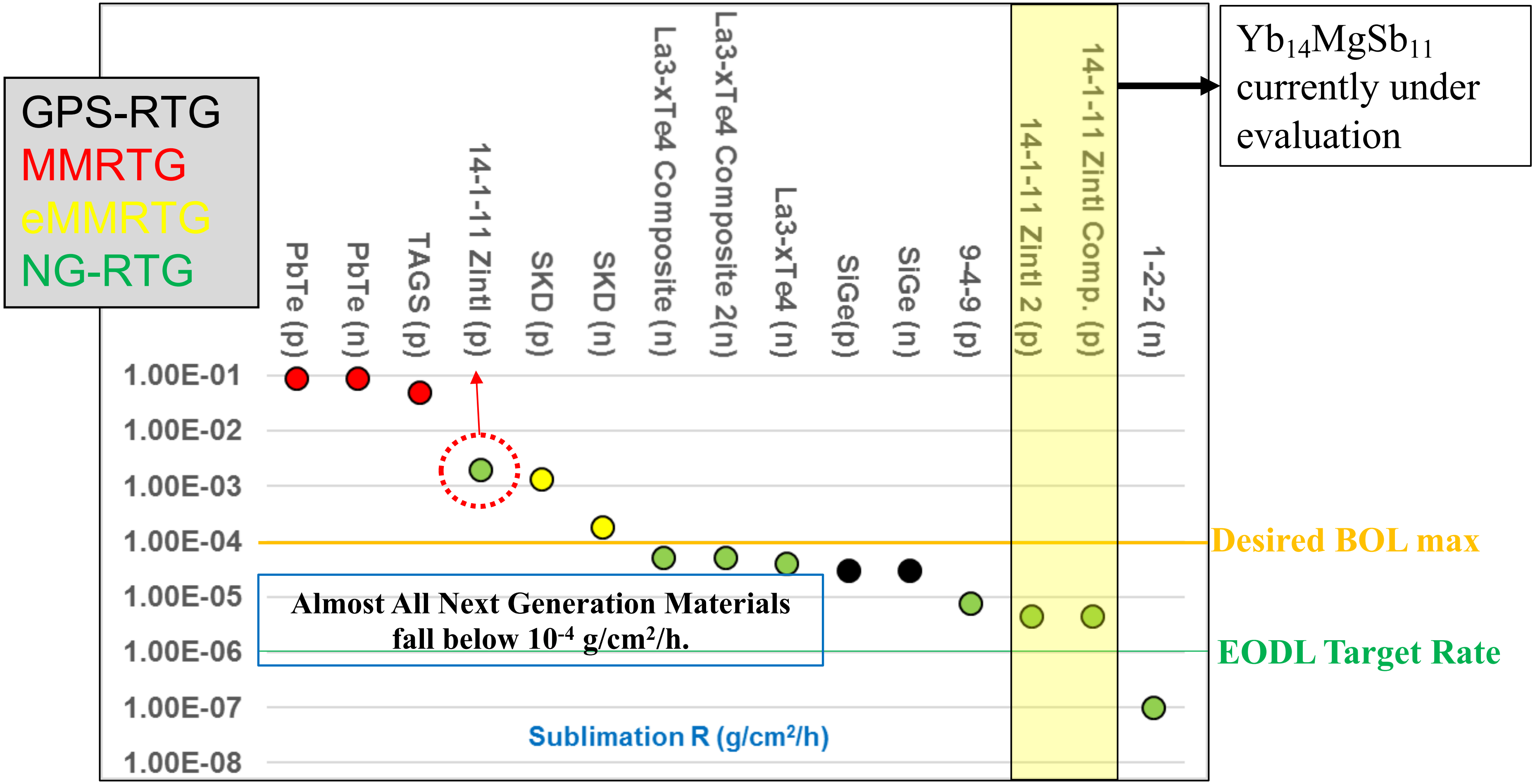


- Substantial increase (>28%) in characteristic strength of $La_{3-x}Te4$ after compositing with Ni.
- Improved mechanical properties lead to higher dicing yield

	n-SKD	p-SKD	$La_{3-x}Te_4$	$La_{3-x}Te_4$ composite	14-1-11 Zintl	n-SiGe
Characteristic strength (MPa)	100	99	35	45	40	192
Weibull Modulus	8	4.4	4	8	4.3	7.4
Number of test samples	8	8	30	10	8	21
Test type	ROR	ROR	ROR	ROR	ROR	4-pt

Sublimation Suppression and Thermal Insulation

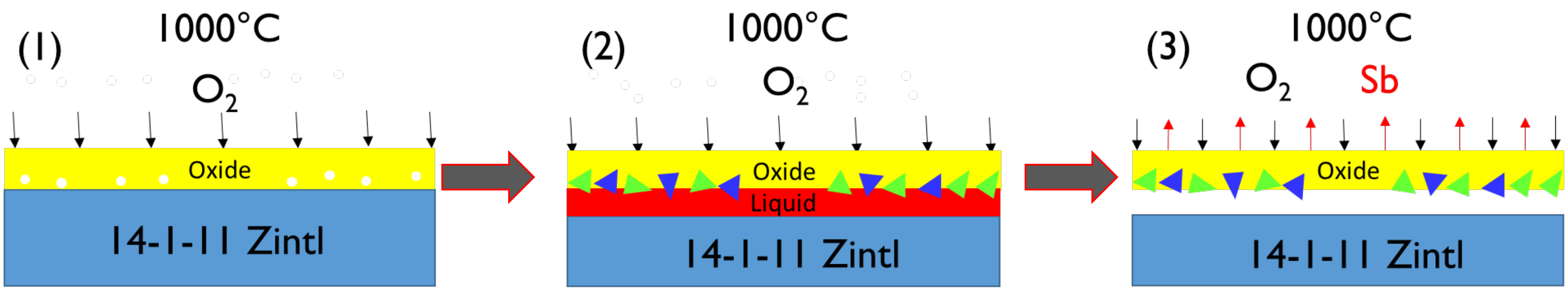
- **Bare Sample BOL Sublimation Rates in Vacuum and Nominal Operating Temperatures**



❑ However, sublimation suppression approaches need to be developed in order to reduce sublimation rates to ~ 10-6 g/cm²/h.

Device Packaging – Integration with Thermal Insulation

- TE are in direct contact with sublimation suppression and thermal insulation components.
- Chemical reactivity of TE materials with metals and oxides in converter can make this a challenging problem:



- TE material in direct contact with porous refractory oxide.

- Oxide reacts with TE material.
- Deviations from TE stoichiometry lead to the formation of liquid phase

- Sublimation of liquid phase leads to continued reactivity.

- High chemical reactivity at high operating temperatures between TE materials and converter components can lead to significant performance degradation over the long design life of the generator.

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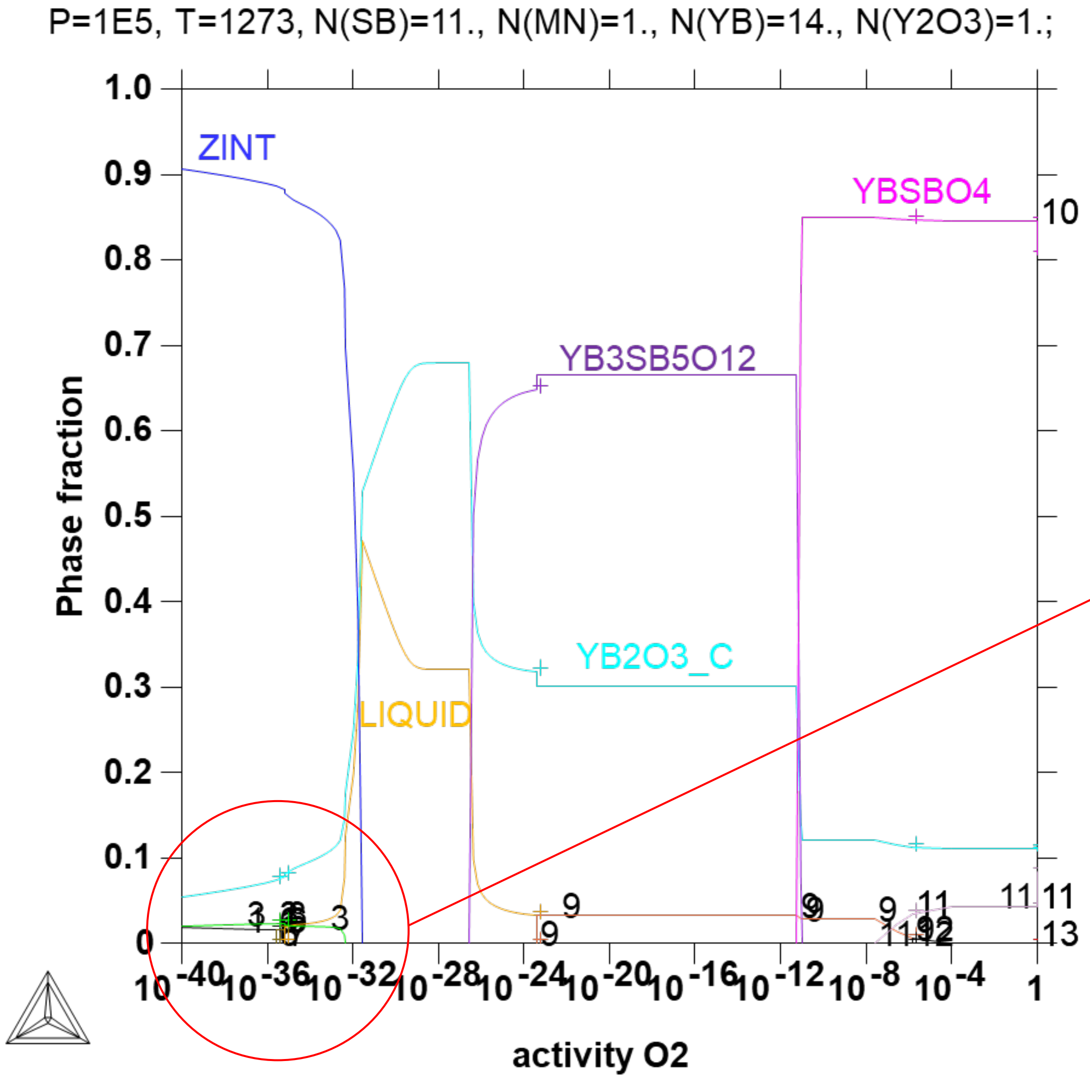
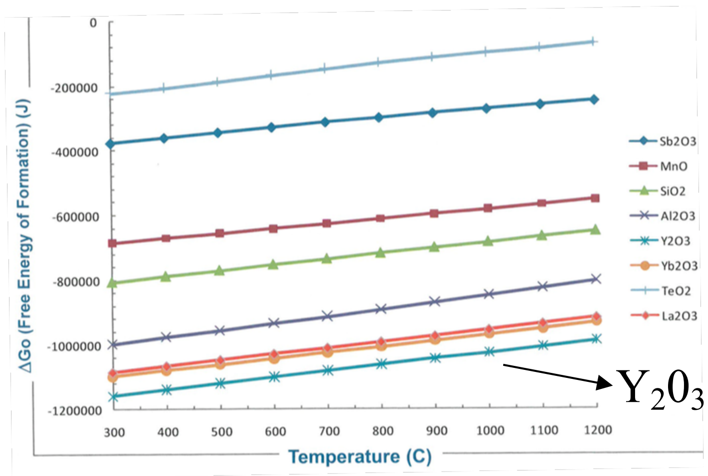
Development of Stable 14-1-11 Zintl/Oxide interfaces

Example: 14-1-11 Zintl to Y_2O_3 Interaction





- Formation phase persists even at extremely low oxygen activity.
- Currently investigating more stable chemistries.

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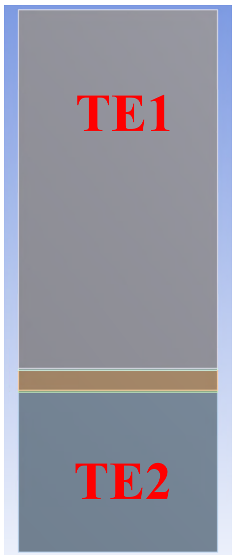
Development of Compliant Interfaces

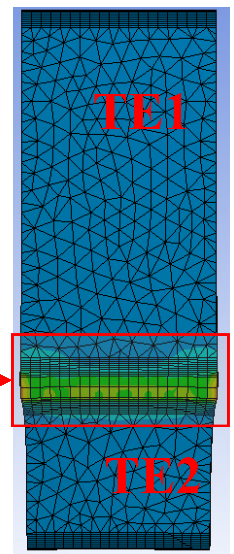
- Additive manufacturing allows for the rapid fabrication of unique structures that are not easily attainable via conventional machining.



Compliant Structure is bonded between the high-temperature (HT) and low temperature (LT) TE sections

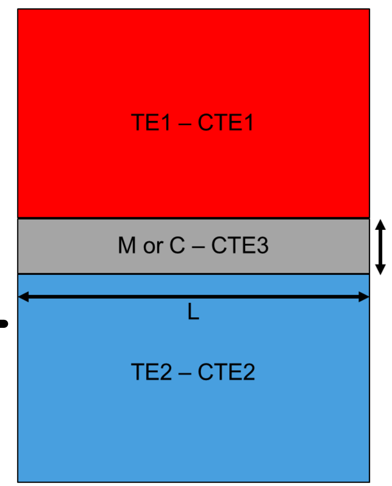
Deformation on cooldown (HT to RT)

Reduced Mechanical Coupling between TE components



During Cooldown, because of CTE mismatch, the LT TE contracts more than the HT TE, which leads to high stress and subsequently crack formation. *The compliant structure mechanically isolates the HT TE form the LT TE Components, hence reduces stress transfer and eliminates crack formation.*

Generic Physical Model



TE: Thermoelectric Element
CTE: Coefficient of Thermal Expansion
M: Metal
C: Compliant

Stress in TEs Is Reduced When

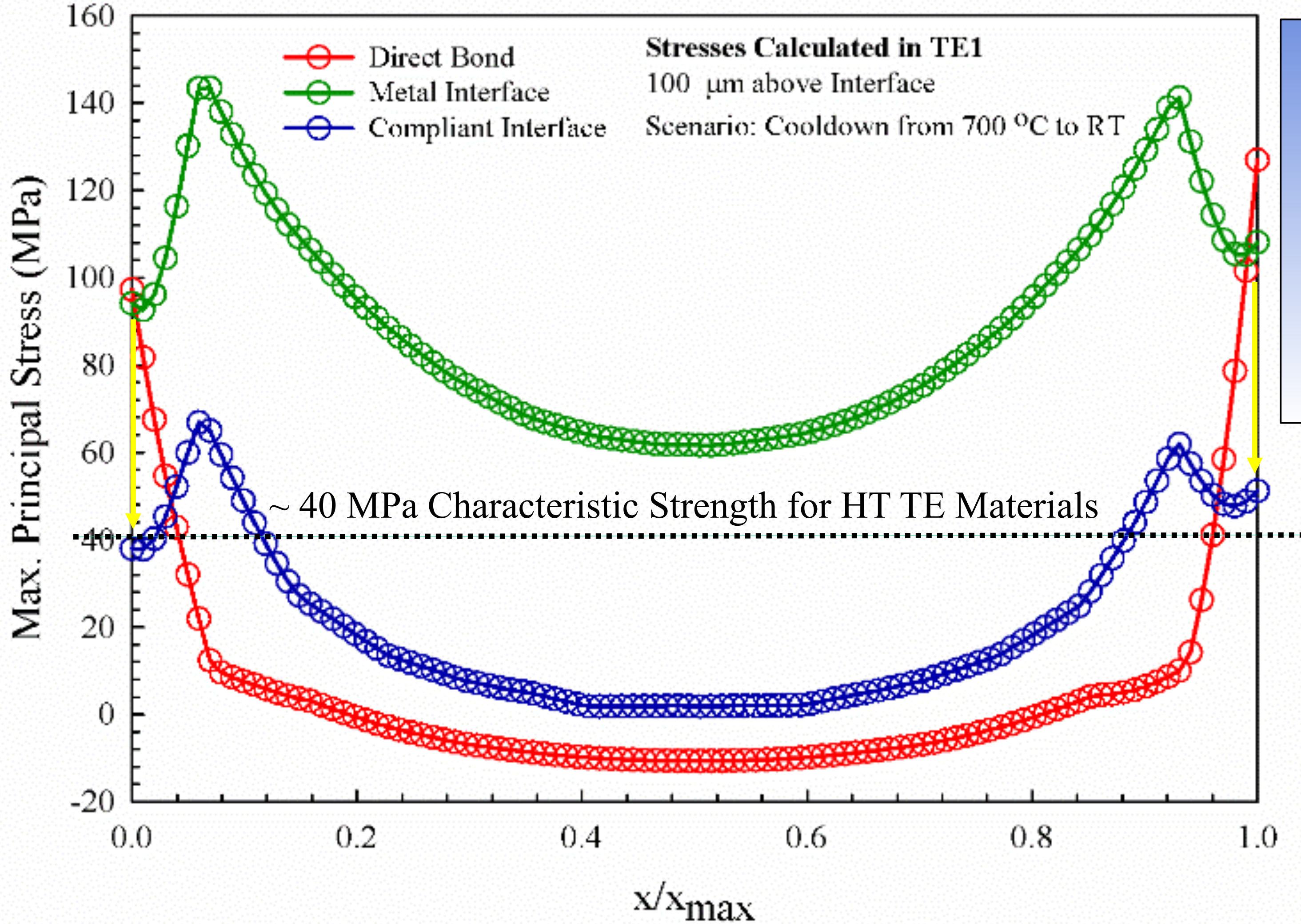
- CTE1 ~ CTE2 ~ CTE3 (CTE Matching)

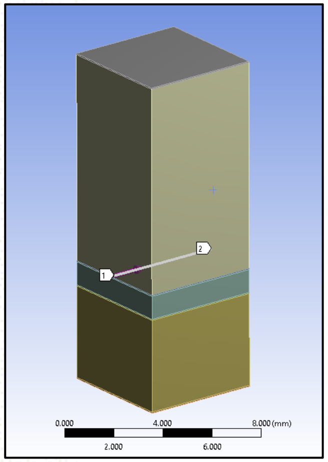
Or

- Presence of Low Modulus Intermediate Layer (Compliant Layer)

- Reduced mechanical coupling is due to the geometric intricacies of the compliant structure.

Development of Compliant Interfaces

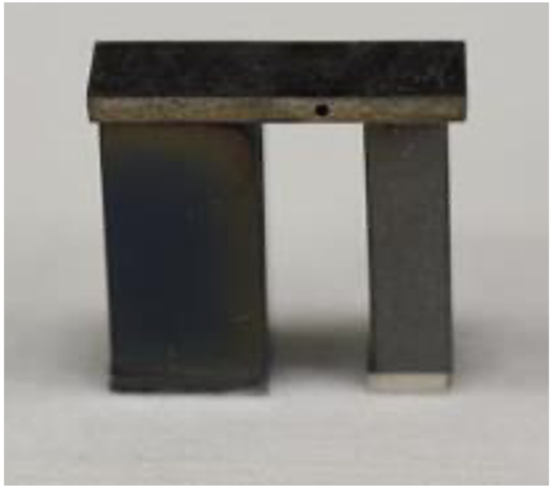




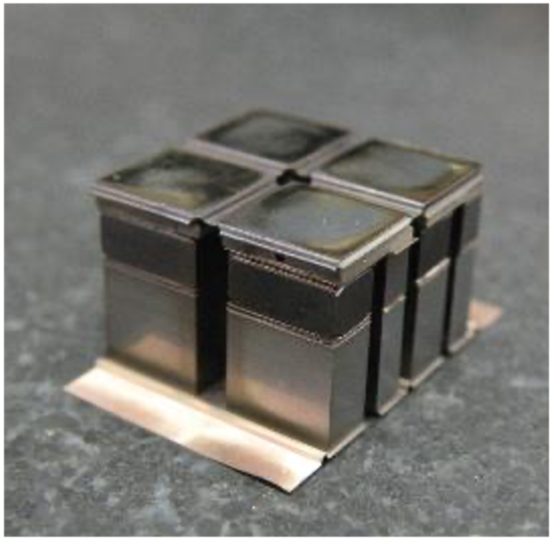
- >50% Reduction in Max Principal Stress at edges

Spring Loaded Devices – Fabrication and Testing

Spring loaded couple



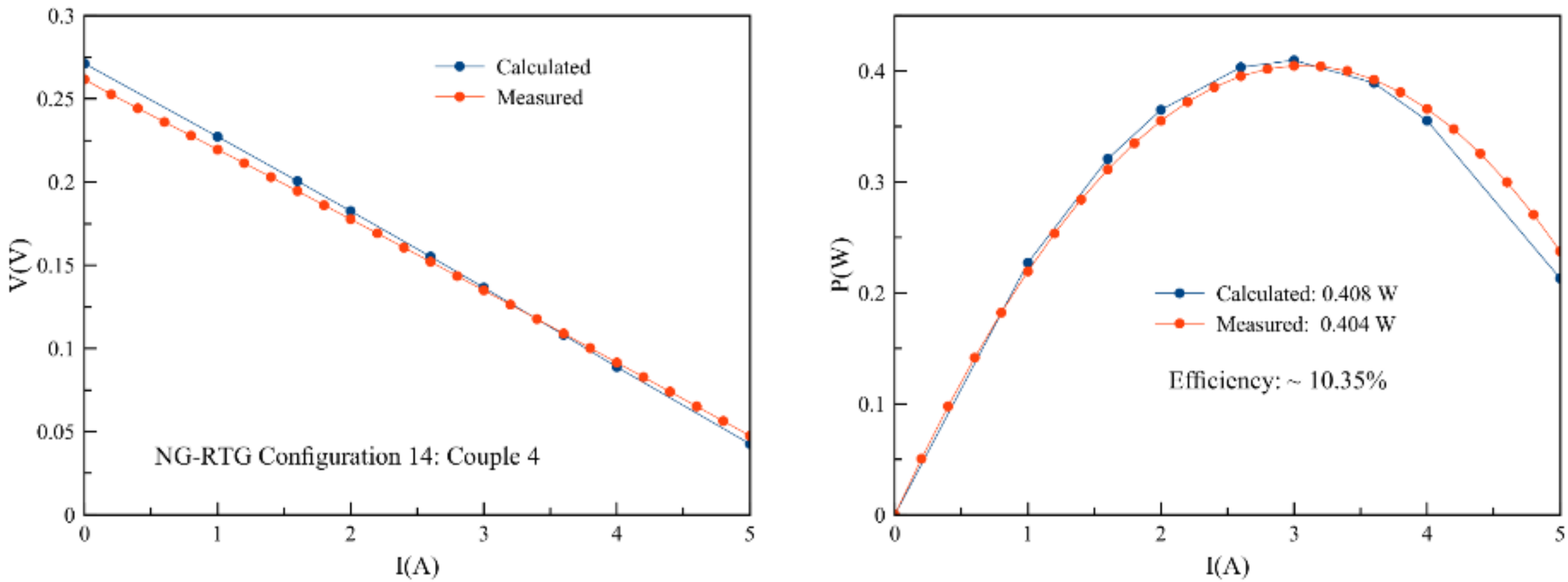
Spring loaded segmented multicouple



- Spring loaded devices are used to validate TE performance, and quickly identify degradation mechanisms.
- Once dominant degradation mechanisms have been identified, appropriate changes to the device configuration are implemented - iterative process designed to eliminate catastrophic failure mechanisms.
- Devices are tested at multiple hot-junction temperatures in order to determine the optimal hot junction temperature (minimum impact on performance and concurrent maximum longevity).

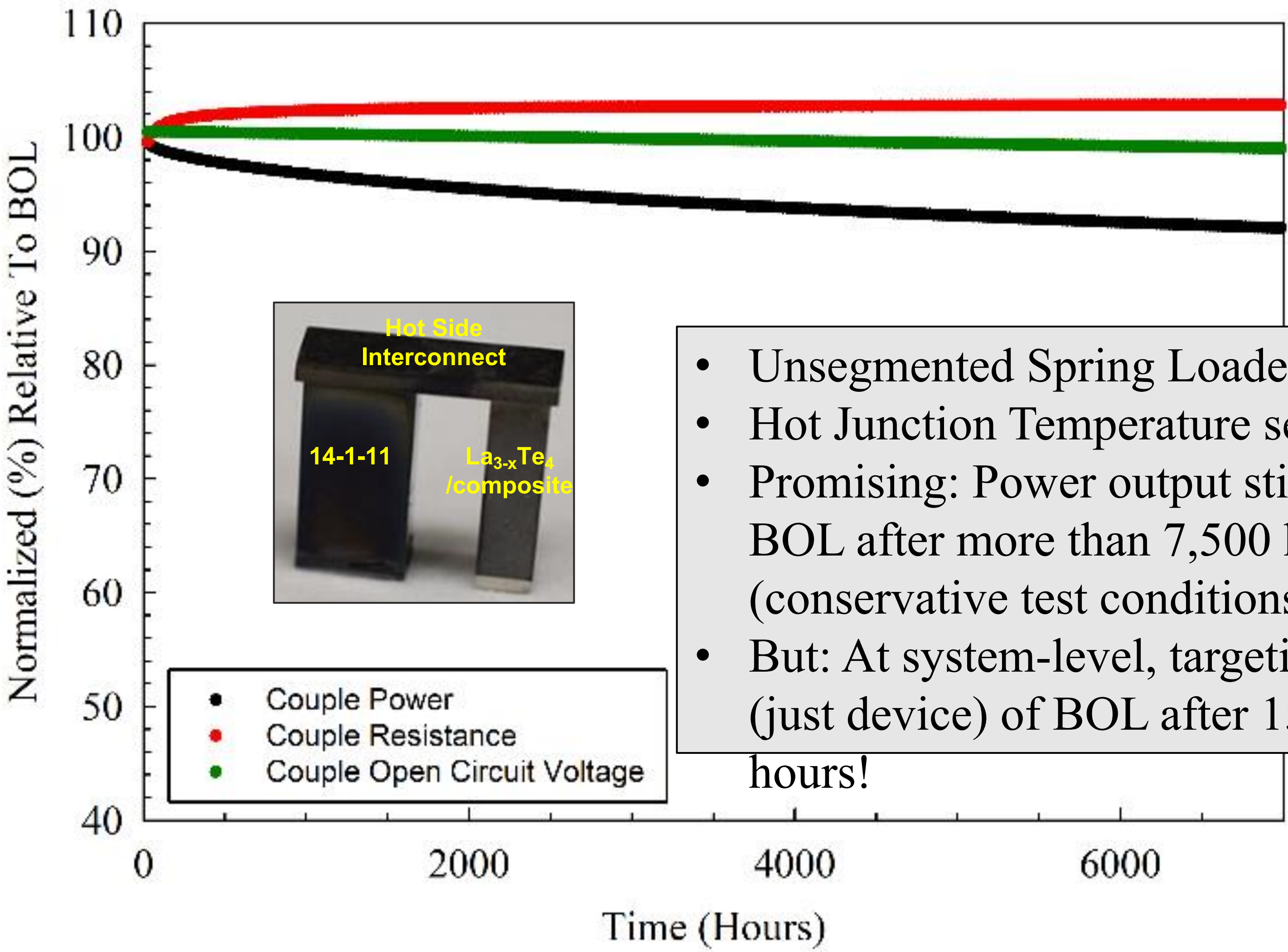
Devices Highlight – BOL Performance Prediction

BOL Data Device Level Verification – Long Term Testing Currently In Progress



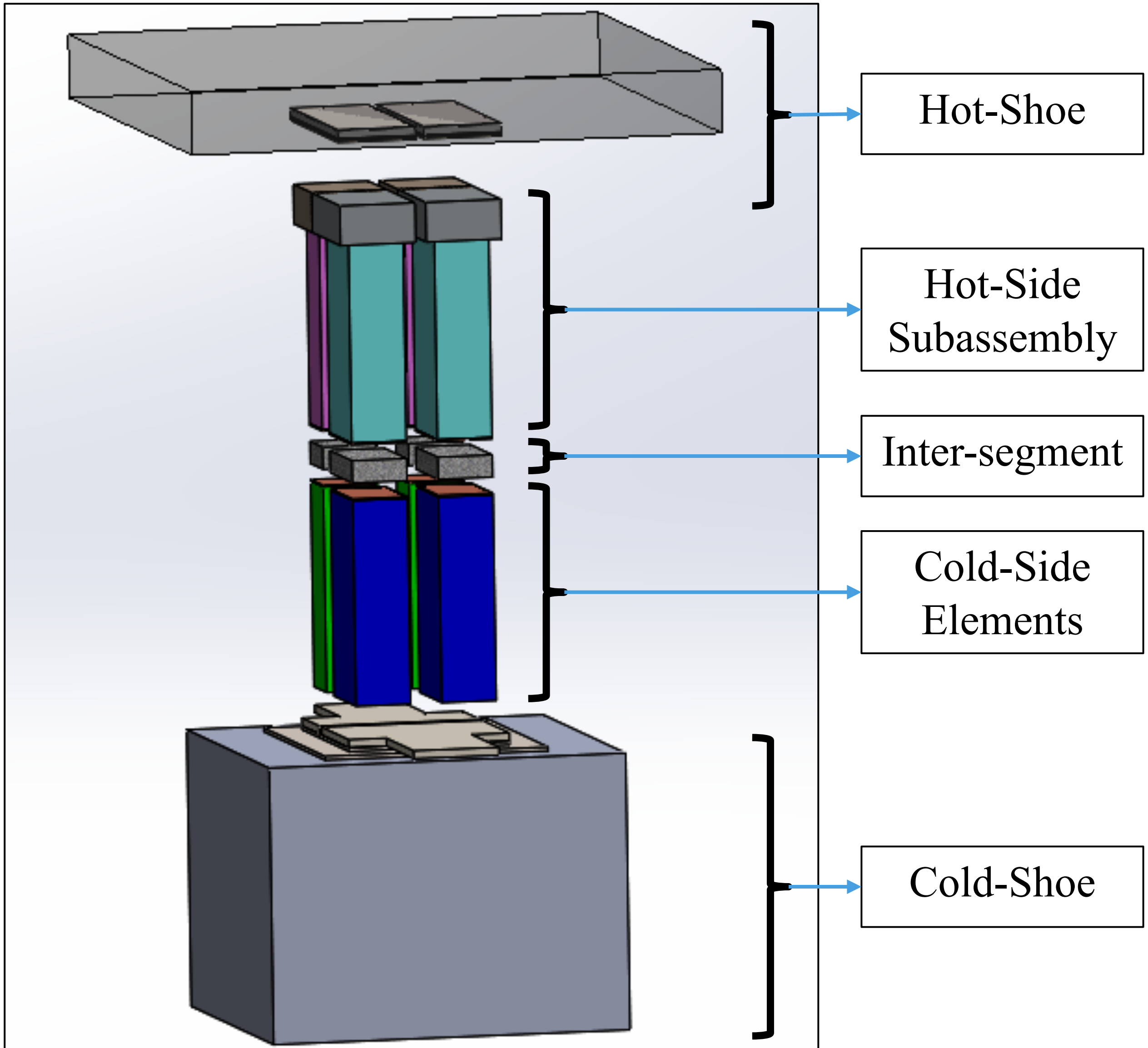
- $T_{hj} \sim 1000\ ^oC$, $T_{cj} \sim 200\ ^oC$
- Good agreement between experimental data and FEA calculations.
- Pmax~ 0.410 Watts @ I=3 A
- Efficiency e ~ 10.35%

Extended Performance Testing

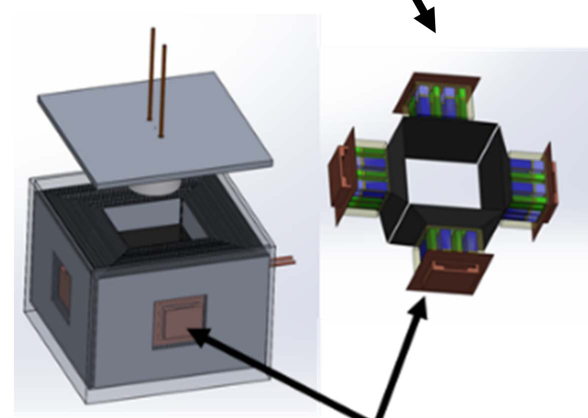


- Unsegmented Spring Loaded Couple
- Hot Junction Temperature set at 900°C
- Promising: Power output still at ~94% of BOL after more than 7,500 hours (conservative test conditions)
- But: At system-level, targeting ~90% (just device) of BOL after 150,000 hours!

Current Focus - Cantilevered Multicouple



- Proof-of-principle CL Multicouples currently being assembled for both segmented and unsegmented configurations
- Sequence of high temperature bonding steps
- Devices will be tested in the already fabricated MLD (Multi-device Life Demonstrator).



Conclusions

- Set of thermoelectric materials and device configurations have been selected for converter technology development and possible infusion into a Next Gen RTG .
  - Scale-up synthesis has been demonstrated for all relevant thermoelectric materials.
  - Considerable progress has been made measuring relevant temperature-dependent material properties (TE Properties, Mechanical Properties, Bare Sublimation Rates).
  - Extended (1-2 years) thermoelectric property stability testing has been completed for several of these materials.
  - Targeting completion of all materials development and characterization work in FY19.

- Converter technology development is in progress
  - Focus is on two segmented and one unsegmented cantilevered multicouple device configuration.
  - Developed compliant interfaces that minimize stress due to CTE mismatch and device assembly steps.
  - Developed FEA models for thermomechanical analysis and thermoelectric performance analysis to help guide design trades.
  - First round of extended device performance testing is underway.

- Selected device configurations offer ample margin against initial Next Gen RTG performance target
  - Will help minimize initial technology development risks by trading performance and operating temperatures.

Acknowledgements

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